EXPERIMENTAL INVESTIGATION OF IN SITU CLEANABLE OR REGENERABLE FILTERS FOR HIGH-LEVEL RADIOACTIVE LIQUID WASTE TANKS

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ABSTRACT

The Westinghouse Savannah River Company, located at the Savannah River Site (SRS) in Aiken, South Carolina, is currently testing two types of filter media for application as in situ regenerable/cleanable filters on high-level radioactive liquid waste tanks. These HLW tanks range in storage capacity from three-quarters of a million gallons to 1.3 million gallons. Each of these tanks is equipped with an exhaust ventilation system to provide tank ventilation and to maintain the tank contents at approximately 1-in. water gauge vacuum. These systems are equipped with conventional, disposable, glass-fiber, high efficiency particulate air (HEPA) filters that require frequent removal, replacement, and disposal. The need for routine replacements is often caused by accelerated filter loading due to the moist operating environment, which structurally weakens the filter media. This is not only costly, but subjects site personnel to radiation exposure and possible contamination.

The types of filter media tested, as part of a Department of Energy, National Energy Technology Laboratory procurement, were sintered metal and monolith ceramic. The media were subjected to a hostile environment to simulate conditions that challenge the tank ventilation systems. The environment promoted rapid filter plugging to maximize the number of filter loading/cleaning cycles that would occur in a specified period of time. The filters were challenged using particles of non-radioactive, simulated high-level waste and atmospheric dust, as these materials are most responsible for filter pluggage in the field. The filters were cleaned/regenerated in situ using an aqueous solution of dilute (10% volume) nitric acid.

The study found that both filter media were insensitive to high humidity or moisture conditions and were easily cleaned in situ. The filters regenerated to approximately clean filter status even after numerous plugging and cleaning cycles. The filters were leak tested using poly-alpha olefin aerosol at the beginning, middle, and end of the test campaign. Both the sintered metal and ceramic filters passed the challenge test with the efficiency of a conventional HEPA filter (≥ 99.97%) each time.

The sintered metal and ceramic filters not only can be cleaned in situ but also hold great potential as a long life alternative to conventional HEPA filters. The Defense Nuclear Facility Safety Board Technical Report, entitled HEPA Filters Used in the Department of Energy's Hazardous Facilities, found that conventional glass fiber HEPA filters are structurally weak and easily damaged by water or fire. The structurally stronger sintered metal and ceramic filter media would reduce the potential of a catastrophic HEPA filter failure due to filter media breakthrough in the process ventilation system. An in situ regenerable system may also be applicable to recovery of

nuclear materials, such as plutonium, collected on glove box exhaust HEPA filters. This innovative approach of the in situ regenerable filtration system may prove to be a significant improvement upon the shortfalls of conventional disposable HEPA filters.

LIST OF ACRONYMS

ATI Air Techniques International

DOE U.S. Department of Energy

DOP di-octyl phthalate

FTF Filter Test Facility

HEPA high efficiency particulate air

HFTA HEPA filter test apparatus

HLW High-level Waste

PAO Poly-alpha olefin

SC South Carolina

SRS Savannah River Site

SRTC Savannah River Technology Center

I. BACKGROUND

Conventional disposable glass-fiber HEPA filter cartridges are used throughout the U.S. Department of Energy (DOE) complex in various process systems. These filters require routine removal, replacement, and disposal. This process is not only expensive, but subjects site personnel to radiation exposure and adds to an ever-growing waste disposal problem. The conventional HEPA filters also have safety concerns regarding the strength of filter media, water damage, and operation in environments with elevated temperatures. The Defense Nuclear Facility Safety Board issued a report entitled "HEPA Filters used in the Department of Energy Hazardous Facilities", DNFSB/TECH-23, in which these and other concerns pertaining to conventional HEPA filters were addressed. (1)

The SRS High Level Waste (HLW) tanks are designed to hold approximately 1.0 million gallons of radioactive liquid waste. Figure 1 depicts a typical HLW tank ventilation flow diagram. The tanks are located outdoors and buried in the ground except for the tank top. They are equipped with a ventilation system that provides approximately 500 cfm of filtered air within the headspace. The airflow maintains the tank contents at negative pressure (-1.0" water column). The air flows through a demister and condenser where water is removed from the air stream. The flow then enter a reheater where the air is heated above dew point before being filtered at the outlet HEPA Filter and released to the atmosphere. Tanks with secondary containment also have a separate ventilation system for the annulus space. The negative pressure maintained on the tank contents prevents the release of radioactive material to the environment. However, it also allows for atmospheric dust to be pulled into the tank during normal operation.

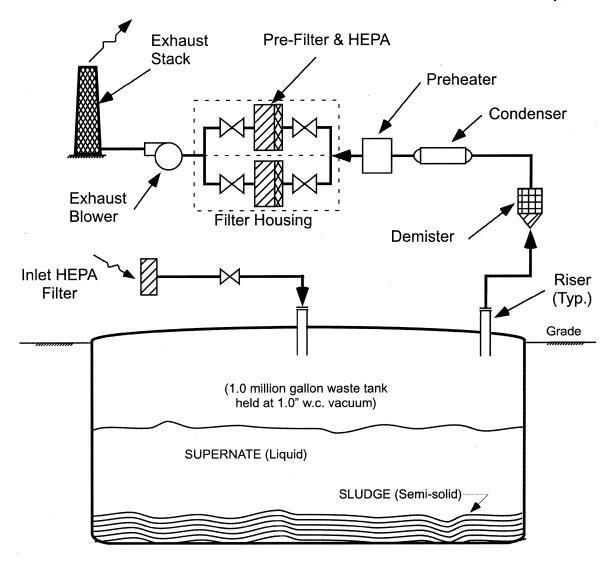


Figure 1: Typical HLW ventilation system flow diagram

The glass fiber HEPA filter must exhibit a particle removal efficiency of 99.97% when challenged by thermally generated poly-alpha olefin (PAO) with a diameter of approximately 0.3 microns. It is common practice at SRS to refer to this test as a DOP test, in reference to di-octyl phthalate that was commonly used in the past as the standard test material in HEPA filtration. The acronym, DOP, will be used throughout the remainder of this report. The pleated glass fiber HEPA filter media has approximately 240 ft² of surface area and is typically contained in a 2 ft x 2 ft x 1 ft housing and exhibits a 1 inch of water column (w.c.) differential pressure (dP) across the filter media when clean. A HEPA filter remains in service until the filter media reaches a predetermined maximum pressure drop (approximately 5 inches w.c.) or a high source term due to radioactive buildup, and then the filter is replaced.

SRS requires that all process HEPA filters pass an in-place leak test both before being placed into service, and periodically thereafter. DOP test connectors are designed into each HEPA installation to facilitate the routine in-place DOP test per national and site standards. Both the material challenge and the filter efficiency tests conducted on the filters in this study were designed to simulate conditions found in HLW tanks at SRS.

Previous research has been conducted at other DOE sites such as Lawrence Livermore National Laboratory and Oak Ridge National Laboratory to develop an in situ cleanable HEPA filter with high media strength. W. Bergman, et al. conducted research on various filter media, such as steel fibers, ceramic, and sintered metal, using reverse air pulse as the in situ cleaning method. ⁽²⁾ The results of these investigations indicate that commercially available filter media could be applied to the development of an in situ cleanable (using reverse air pulse) HEPA filter system that would meet the performance criteria established for a conventional HEPA filter system.

In FY98 SRS started working on the theory of using high-pressure drop filter media in an in situ regenerative filtration system. The in situ cleaning of the filter media used an aqueous solution to regenerate the media to a new filter status. Testing conducted in FY98 and FY99 indicated that an in situ cleanable filtration system is feasible for use on the HLW tanks.

A cost analysis conducted on cleanable HEPA filters showed that the in situ cleanable filters have the potential of being cost effective when compared to the conventional glass fiber HEPA filters. The study estimated that the DOE complex uses an average of 11,000 filters per year at an estimated cost of \$55 million. Using in situ cleanable HEPA filters could save the complex \$42 million a year in operating cost. The study did not include costs associated with glass fiber filter breakthrough or rupture, but these additional costs increase the cost benefit of the in situ cleanable filters for the complex. (2)

II. INTRODUCTION

The DOE Tanks Focus Area and The National Energy Technology Laboratory (formerly known as the Federal Energy Technology Center) issued a "Call for Proposals" to identify vendors conducting research in the area of in situ cleanable/regenerative filters or vendors interested in pursuing such technology. (3) A technical evaluation was conducted on the proposals.

Based on the proposals reviewed, two filter vendors were selected to support this research program. CeraMem Corporation, located in Waltham, Massachusetts and Mott Corporation, located in Farmington Connecticut.

CeraMem Corporation

A picture of the CeraMem filter (H-18) tested at the Savannah River Technology Center (SRTC) is shown in Figure 2.

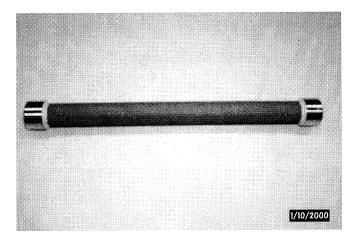


Figure 2: Ceramic test filter

Figure 3 depicts the design of the CeraMem filter media and the airflow passage through the filter. As shown, the dirty air with particulate matter enters the filter media. The entrained particles are then filtered from the air in the microporous membrane. The clean air flows down the clean channels and is exhausted from the filter.

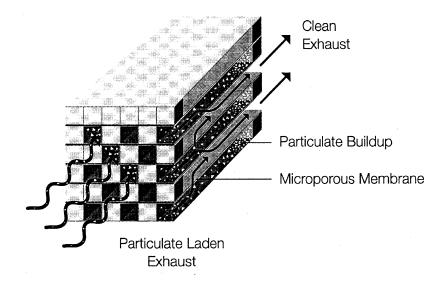


Figure 3: Airflow through ceramic filter media

Below are the characteristics of the ceramic monolith filter tested in this program

•	Monolith Membrane Support:	Silicon carbide
•	Membrane Coating:	Glass-frit-bonded zirconium silicate and alumina
•	End Ring Adhesive and Plugs:	Zirconium-silicate-filled food-grade epoxy
•	End Rings:	316L stainless steel
•	Overall Element Dimensions:	1.28" diameter (end ring) x 13.1" long
•	Ceramic Component Dimensions:	1.06" diameter x 12.0" long
•	Filter Configuration:	Flow-through, dead-ended filter
•	Number of Active Inlet Channels:	30
•	Channel Opening Size:	0.066"
•	Channel Wall Thickness:	0.03"
•	Channel Plug Depth:	0.12"
• "	Inactive Channel Sides:	16 (next to skin around circumference)

Pressure Differential

75" w.c. @ 9 scfm

• Filtration Surface Area:

 0.56 ft^2

A literature review found that previous work had been conducted on an earlier version of a CeraMem honeycomb ceramic monolith filter as an in situ cleanable filter using reverse air pulse. (2) The ceramic filter was not recommended by the study as a cleanable (using reverse air pulse) HEPA filter due to high-pressure drop and particle removal efficiency less than 99.97%.

Mott Corporation

The sintered stainless steel filter provided by Mott is shown in Figure 4. The filter medium was constructed of 1-micron 316L stainless steel particles sintered into a 2 inches diameter by 4 inches long cylindrical geometry and had neck-down tubing that was 0.75 inch in diameter.



Figure 4: Sintered stainless steel test filter

Below are the Mott Test Filter Characteristics:

• Filter Media sintered powdered stainless steel

Overall Element Dimensions: 2.0" diameter x 3.75" long

• Wall Thickness: 0.047"

• Filter Configuration: Flow outside to inside

Pressure Drop 160" w.c. at 9 scfm

Filtration Surface Area: 0.16 ft²

Previous research was also conducted on the Mott sintered metal filter as a potential air backpulse in situ cleanable HEPA filter. The sintered metal was not recommended by the study due to pressure drop across the filter greatly exceeding the 1-inch w.c. requirement of the DOE Standard on HEPA filters and ASME AG-1. (4,5)

III. TEST LOOP DESCRIPTION

A HEPA filter test apparatus (HFTA) was designed and constructed to test the filters. The HFTA was designed to simulate the conditions found on the HLW tanks at SRS. Figure 5 depicts the schematic of the HFTA constructed at the Thermal Fluids Laboratory at SRTC.

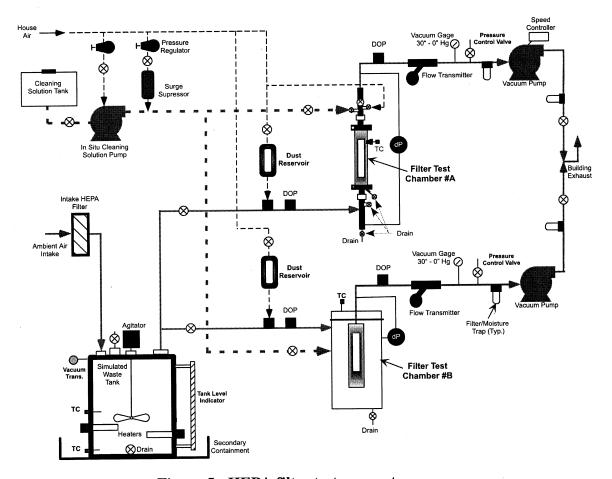


Figure 5: HEPA filter test apparatus

The test was designed for approximately 18 scfm of filtered air to flow into a simulated waste tank filled with approximately 30 gallons of heated (104° F - 140° F) simulated HLW solution. The simulant solution was mechanically agitated while the filtered air traveled through the headspace of the tank. This flow path simulated the HLW tank ventilation system. After exiting the tank, the airflow was split between the CeraMem and Mott filters, with 9 scfm of measured air passing through each filter. A separate vacuum pump was used to pull the air across each filter as shown in figure. DOP test connectors were designed into the HFTA to conduct in-place leak tests on the filter media.

The HFTA was designed such that one filter could be cleaned while the other remained online for testing. The system operated continuously (24 hours a day, 7 days a week) until the flow across the filter(s) decreased by 20% or more due to particulate matter build up on the surface of the filter. The time to plug a filter was determined by many variables such as test solution, speed of the agitator, liquid level in the tank, etc. Once a filter became soiled from the simulated operation, it was cleaned using the in situ cleaning system shown in Figure 5. Spraying the inlet side of the

filter with 10% (by volume) nitric acid, rinsing with water, then spraying with a 10% sodium hydroxide solution followed by a final rinsing with water cleaned the Mott filter. After the in situ wash cycle, the filter was returned to operation. The CeraMem filter was initially cleaned in situ by injecting a solution of a 10% nitric acid into the dirty side of the filter and allowing the filter to soak. The filter was then back flushed with water. This method was found to be an inefficient cleaning method. Therefore, the in situ cleaning method was modified such that a 10% nitric acid solution was back-flushed through the filter and allowed to soak in the filter for approximately 10 minutes. The filter was then back-flushed with water. This modified cleaning method improved the cleaning efficiency of the CeraMem filter.

IV. TEST SOLUTIONS

Filtering entrained particulates of simulated HLW and atmospheric dust in the air stream challenged the filters. These materials, neglecting the radioactive constituents, are believed to be responsible for plugging the existing HEPA filters in the HLW tanks, especially under high humidity conditions found in the tanks. Two solutions were mixed to simulate the HLW found in HLW tanks at SRS. Only one solution was in the waste tank at a time to challenge the filters. The formulations for the HLW simulant constituents are shown below.

Simulated HLW Sludge				
Element	wt %	Element	wt %	
Al	17.66	Ba	0.40	
Ca	2.32	Cr	0.24	
Cu	0.20	Fe	31.76	
Mg	0.26	Mn	10.50	
Na	21.73	Nd	3.42	
Ni	2.10	Pb	0.48	
Sr	0.70	Zn	0.33	
Zr	0.10	K	0.36	
Cs	0.02	Si	7.42	

HLW Simulated Salt

- 8.0 M NaOH (Sodium Hydroxide)
- 1.5 M NaNO₃ (Sodium Nitrate)
- 1.7 M NaNO₂ (Sodium Nitrite)
- 0.4 M NaAl(OH)₄ (Sodium Aluminate)

South Carolina Road Dust

South Carolina road dust was used to simulate atmospheric dust around SRS. The SC dust consisted of topsoil from Aiken County. The soil was dried in an oven to remove the moisture and particles larger than 75 microns were sifted out. The dust was slowly injected from the dust reservoir into the 9-scfm air stream using a small stream of air. The simulated waste tank was empty and the air temperature was approximately 70° F.

V. RESULTS AND DISCUSSION

• The CeraMem and Mott test filters passed the in place leak DOP test with 99.97% particle removal efficiency at the beginning, middle and end of the test campaign.

- Additional filters from Mott and CeraMem were DOP tested and two Mott filters failed the efficiency test. This is of concern since approximately 19 elements will be used in a full-scale system for a HLW tank.
- The Mott filter operated in a high humidity environment with little or no effect on its operation.
- The performance of the CeraMem filter was adversely affected during the high humidity test conditions. However, the design of the filter housing may have caused the poor performance.
- The CeraMem and Mott filters both regenerated well during the in situ cleaning. That is, the airflow and dP across the filters returned to a clean filter status after being challenged with simulated HLW sludge, South Carolina road dust and simulated HLW salt. (6,7)

The first performance test on the CeraMem and Mott filters was a standard in-place leak (DOP) test of HEPA filters, conducted by the site HEPA Filter Test Group at SRS. Both the CeraMem (H-18) and Mott (36699-01-00-2225079-010, 11/97) test filters passed the leak test with a 99.97% removal efficiency or greater. The same test filters were also DOP tested after the SC road dust test and at the end of the test campaign. Both test filters had an efficiency of 99.97% or greater each time. This proved that the filter media was not physically breaking down due to the simulants or the in situ cleaning during the test campaign.

Three additional Mott filters and one CeraMem filter were also in-place leak tested. The CeraMem, H-19 filter passed the DOP tested with greater than 99.97% efficiency. However, two of the three Mott filters failed the DOP test. Mott Filter 2225372 #1 (0.028" media) failed the filter challenge test with a 99.95% efficiency. Mott Filter 2225372 #2 (0.028" media) passed the filter challenge test with a 99.99% efficiency. Mott filter PN202-01-00 2225079-010, #1, 7/99 (0.047" media) failed the test with only a 99.00% efficiency.

It was suggested that the filter failure might be due to the filter media imperfections near the welded surface. Figure 6 presents electron-microscope pictures of the failed sintered media (Mott filter 2225372 #1). The picture on the left depicts the suspected area likely causing the failure. This area is between the weld and the normal filter media. The picture on the right is what a typical sintered stainless steel filter media looks like when magnified 500x. However, it cannot be proven with certainty that the welded area caused the filter failure.

Concerns exist also on using organic challenge materials (PAO or DOP) for these alternative media. Small particle sodium chloride may be a better challenge material for this media. Sodium chloride testing has been used with great success in Great Britain and Continental Europe for the last 50 years. (8) A study needs to be conducted to optimize the method and challenge material for leak testing (DOP) the alternative media.





Figure 6: Electron micrograph of the sintered stainless steel media

The CeraMem and Mott filters were tested with water in the simulated waste tank to determine if the filters could operate in a high humidity environment without operational problems. The water in the waste tank was agitated and heated until the humidity was approximately 100% in the filter housings. The sintered metal filter performed well in the high moisture air stream. However, the ceramic filter was blinded during the high moisture conditions. This poor performance of the ceramic filter may have been a result of the design of the filter housing which allowed water to condense and collect on the filter media, in turn, blinding the filter.

While challenging the filters with the simulated HLW sludge, both filters performed well overall. The Mott filter did not see complete recovery or regeneration after each in situ cleaning during the sludge test. The airflow across the filter degraded from 9 scfm to approximately 7.5 scfm. However, by the end of the test campaign, the flow across the filter had recovered to the original 9 scfm. This degradation of the Mott filter during the simulated sludge test was not seen during the earlier FY98 testing. (9) The CeraMem filter was affected by the high humidity. The system was modified so that condensed water would drain from the housing to prevent blinding of the filter. The CeraMem filter flow increased from 9 scfm at the beginning of the test campaign to 10 scfm. An analysis of the spent cleaning solution found that Al_2O_3 (aluminum oxide) was removed during the cleaning cycle. The Al_2O_3 residual was left on the filter from a turbidity test conducted by CeraMem before shipping the filters to SRS.

The second test was conducted using South Carolina road dust to simulate atmospheric dust inleakage that may occur on the HLW tanks. The dust was slowly injected into the air stream using a small purge of house air. The airflow and dP across the Mott filter recovered after each in situ cleaning cycle. Figure 7 shows the results of loading and cleaning the sintered stainless steel filter in situ. The curves are typical results for the ceramic filter also.

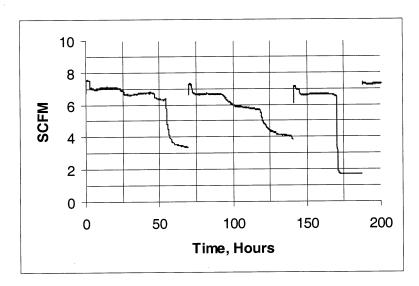


Figure 7: Filter loading using SC road dust and cleaning cycles

Unlike the simulated salt test described below, the SC road dust was quite visible on the surface of the filters. Even with visible dust in the filter housing and on the surface of the Mott filter, it did not deadhead (no airflow across the filter) during a heavy loading of dust on the filter. However, the CeraMem filter was easily deadheaded with the dust. This rapid plugging may also be due to the filter and filter housing configuration. The configuration of the CeraMem filter did not allow

for dust particles to fall out in the filter housing, as seen with the Mott filter/filter-housing configuration. The ceramic filter completely recovered during the in situ cleaning. This condition of filter plugging is a worst case scenario, where the vacuum pump was allowed to operate deadheaded for several hours allowing the small dust particles to be pulled into the filter's flow channels, potentially making it more difficult to regenerate the filter in situ.

The Mott and CeraMem filters also performed well during the salt test. After each in situ cleaning cycle, the airflow/dP across the Mott and CeraMem filters recovered completely. As with the FY98 testing, no buildup of salt particles could be seen on the surface of the Mott filter by visual inspection through the transparent top of the filter housing. The solid stainless steel housing of the CeraMem did not allow for visual inspections.

The ceramic and sintered metal filters operated in a controlled but very hostile environment so that the filters would plug in a relatively short period of time. The filters were plugged and cleaned in situ many times with the airflow and dP across the filter completely recovering over the entire test campaign.

The unknown quantity of radionuclides that may accumulate on a HEPA filter is a concern in the case of a catastrophic HEPA filter failure. Application of in situ cleanable filters could potentially prevent the buildup of alpha and beta emitters. The in situ cleanable filter could also be used in other applications such as to permit recovery of nuclear materials that collect on HEPA filters. Current design concepts for the system emphasize welded metal construction that will eliminate components prone to fail under fire or other high temperature conditions. Heavy smoke and smoke borne particulates may plug the filter, but test results indicate that the plugged condition will not cause filter failure and breakthrough and that in situ regeneration would be feasible.

Even though the filters have demonstrated great potential as an in situ cleanable HEPA filter, much work will be required to certify the filters per the requirement of Code on Nuclear Air and Gas Treatment, ASME AG-1. Work is ongoing with Air Techniques International (ATI) located in Baltimore, Maryland to conduct efficiency testing of the alternative filter media. ATI also operates the DOE Filter Test Facility (FTF) at Oak Ridge. The FTF is responsible for conducting leak tests on conventional HEPA filters before installation at DOE facilities. ATI is in the process of making modifications to their test equipment to conduct efficiency test on the alternative filter media (sintered stainless steel and ceramic). This and other related certification issues will have to be resolved before full-scale commercial replacement of conventional disposable HEPA filters with a new generation of permanent, cleanable HEPA filters, can move forward.

We have shown the feasibility of developing an in situ cleanable filter system. However, the small-scale laboratory work must be scaled up to full-scale equivalents. This represents the next step in the development of the in situ cleanable HEPA filter for HLW tanks, which is discussed briefly below.

VI. FULL SCALE SINGLE FILTER ELEMENTS

Work is ongoing proving this technology of regenerating filters in situ using an aqueous solution. Mott and CeraMem corporations have designed full-scale filter elements. They are in the process of fabricating full-scale filter elements for testing at SRTC. The test apparatus has been designed and constructed to challenge a single full-scale filter element with HLW simulants. The full-scale test elements will undergo an efficiency test by ATI at the Oak Ridge FTF before simulant testing at SRS. The test will ensure 99.97% efficiency when challenged with a 0.3-micron particle.

Figure 8 shows two full-scale ceramic media. The filter is 12 inches long and 5.66 inches in diameter with approximately 20-ft2 surface area. The media has a pressure differential of approximately 20" we at 10 ft/min.

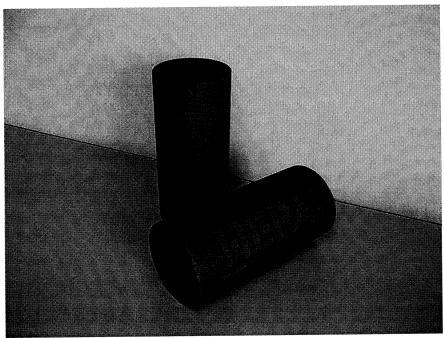


Figure 8: Full-scale ceramic filter element

The full-scale sintered stainless steel filter is depicted in Figure 9. The filter is 24 inches long and 3 inches in diameter with approximately 1.6 ft² surface area. The media has a pressure differential of approximately 42" we at 17 ft/min.

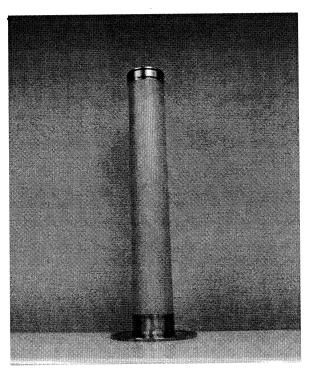


Figure 9: Full-scale sintered stainless steel filter element

VII. ALTERNATIVE FILTRATION FULL SCALE SYSTEM DEPLOYMENT

A full-scale Alternative Filtration System is planned for construction with the hot demonstration of the system being conducted at SRS on HLW Tank 11. HLW tank 11 is a radioactive waste tank containing approximately 0.5 million gallons of radioactive waste. This is a waste removal tank that is scheduled for clean out and final closure in the near future. Before the hot deployment, the full-scale system will undergo cold testing at the SRTC in building 752-25A. SRTC will be the principal investigator on the testing and research of the full-scale system. The Mott filter housing layout being considered for the full-scale demonstration is shown below. Figure 10 is an assembly drawing of the full scale Mott housing. The housing will consist of approximately 19 filter elements as depicted in Figure 11. As currently conceived air will be drawn into the open bottom of each filter element and be pulled through the filter wall by vacuum applied to the clean plenum. The dirty side of the filter will be the inter diameter of the cylindrical element. The multiple vertical tubes will be welded into an arrangement resembling a tube and sheet heat exchanger. After the filter becomes plugged with particulate or when the radioactivity from the accumulated particulate approaches area limits, the in situ cleaning system will be initiated. Each element will have a separate spray nozzle and the filter will be cleaned via spraying the inlet side of the filter with an aqueous solution.

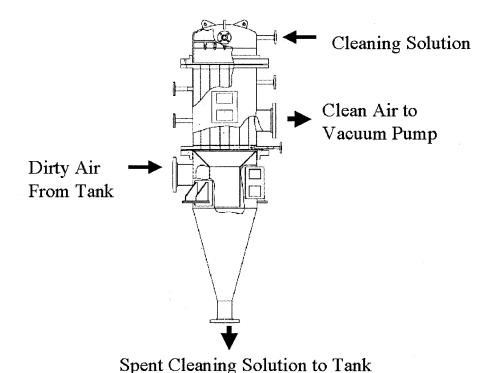


Figure 10: Full-scale filter housing assembly for sintered metal filters

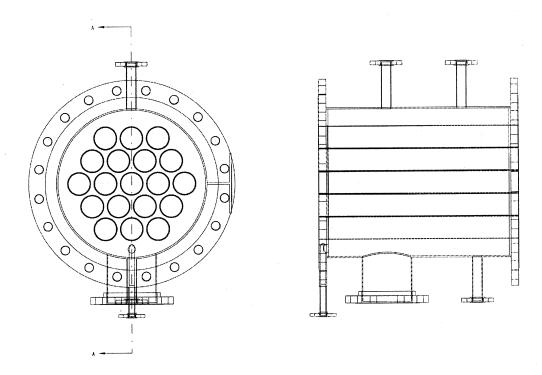


Figure 11: Filter bundle depicting element arrangement

Figure 12 is the conceived CeraMem filter housing for the ceramic media, which is being considered for the full-scale demonstration. The housing will consist of approximately 7 full-scale filter elements as depicted in Figure 13. The dirty air will be drawn into the bottom end of the housing and clean air will exit through the top of housing. The elements are cleaned in situ via back flushing an aqueous solution through the media.

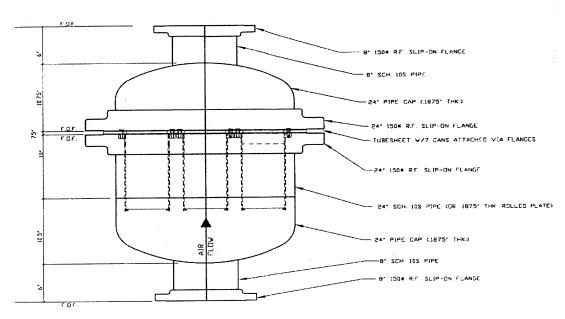


Figure 12: Ceramic full-scale filter housing

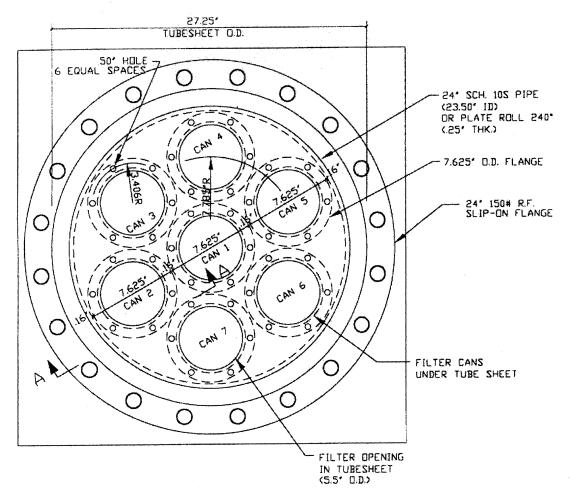


Figure 13: Filter bundle depicting element arrangement

VIII. CONCLUSIONS

Alternatives media such as sintered stainless steel and ceramic membrane holds great promise for use as in situ cleanable/regenerative HEPA filters. The proof-of-principal testing has shown that both ceramic and sintered metal media could be suitable as an alternative HEPA filter media. During simulated testing the filters regenerated well in situ with a potential for 15 year plus life under actual field conditions of the HLW tanks. In addition to eliminating the costs associated with disposing of and replacing disposable filters, these strong filter media also reduce the potential of a catastrophic HEPA filter failure due to rupture of the filter media.

The sintered stainless steel and ceramic membrane filters were plugged and cleaned insitu many times. Each time the differential pressure and flow recovered to a clean filter status. The ceramic and sinter steel filters passed the standard in-place leak test of HEPA filters with an efficiency of 99.97% or better at the start, middle, and end of the test campaign.

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